



The role of time in activation of flow units in metallic glasses



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ABSTRACT

Flow units, which accommodate deformations and initiate transformation from glass to supercooled liquid state, have been proved to significantly influence the properties of metallic glasses (MGs). We study the time dependent activation of the flow units in MGs, and find that the size and the fraction of the flow units increase with the applied time, which lead to the time dependences of the mechanical behaviors, flow phenomenon and relaxations of MGs. A diagram for the flow in glass based on the concept of flow unit, activation time, and imposed energy (stress or temperature) is constructed to understand the deformations and flow mechanisms of MGs.

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1. Introduction

Substances can be divided into solids, liquids and gases based on their macroscopic properties. However, the pitch drop experiment [1] by Parnell has shown that some solids and liquids are hard to be distinguished through their appearance. So another classification, which distinguishes the substances by the orders in their microstructures, is proposed [2,3]. Crystals are the substances with both short-range and long-range orders, while gases have neither of them. The liquids and glasses, which can be called amorphous substances, do not have long-range orders but have short range orders, and this leads to some similar features of glasses and liquids [2].

Unlike freezing or boiling, the transition from liquid to glass has no substantially detectable change in structure. Although it is still disputed whether the glass transition is a slow kinetics phenomenon or a thermal precursor of an obscured “ideal” phase transition [3], experimental results [4] have shown that the glass transition temperature (T_g) is time dependent. Besides, experiments and simulations [5–7] have shown that the slow flow features and mechanical properties of a glass is also time dependent. When a glass is deformed with a low enough strain rate, the glass can behave like a Newtonian liquid [5,6]; while with a very high strain rate, liquid can show fracture behavior similar to that of brittle glass [5].

Metallic glasses (MGs) are the simple atomic glasses among the wide spectrum of glass-forming systems [7–9], because their random disordered structure can be considered as the dense random packing of spheres [10]. The MGs then offer a model system to study the structural origin of flow behavior, deformations, and the glass transition [12–16]. Recently, experimental and simulation evidences [17–27] have shown that the deformation behavior and the mechanical properties of MGs are determined by the intrinsic heterogeneous structure, especially the liquid like regions or flow units in MGs. The flow units play a crucial role in the elastic and plastic deformations, relaxations, rejuvenation, and mechanical and physical properties of MGs. For the features and properties of MGs are time dependent, it is supposed that the flow units are also dependent on time. In this paper, we attempt to characterize the time dependent feature of the flow units in MGs. We find that the activation process, size and distribution of the flow units in MG are indeed time dependent, which is in accordance with experimental results of time dependent macroscopic properties of MGs. A time-relevant diagram for the flow in metallic glasses is proposed.

2. Theory and model

The microstructure of MGs is heterogeneous with nano-scale localized liquid-like sites called flow units embedded in elastic matrix, which strongly affect the features and properties of the glasses [18,22,23]. When stress or temperature is applied, the flow units in MGs will be activated, dissipate the applied energy and flow inelastically. According to the elastic model [9], the activation

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energy W_i of a flow unit in a glass is mainly elastic energy, and is controlled dominantly by its shear modulus G_i [9,28]:

$$W_i = aG_i\Omega_i, \quad (1)$$

where a and Ω_i are the constant coefficient and volume of the flow unit, respectively. The activation energy and activation time have an Arrhenius relation [9]: $t = \tau_i \exp(W_i/kT)$, or $W_i = kT \ln t/\tau_i$, where T is temperature, k is the Boltzmann constant, and τ_i is the intrinsic relaxation time of the flow unit. Therefore, the relation between activation time and Ω_i is:

$$\Omega_i = \frac{kT}{aG_i} \ln \frac{t}{\tau_i}. \quad (2)$$

Eq. (2) indicates that the flow units are not activated spontaneously because the activation process needs sufficient time, and the larger (smaller) flow units with longer (shorter) intrinsic relaxation times need longer (shorter) time to be activated. In other words, the dynamic and energy distribution of the atoms in a flow unit are inhomogeneous, and the flow unit is actually activated gradually: the softest region in a flow unit will be activated first and the harder ones later. Fig. 1(a) is a schematic illustration of a flow unit in MG. The blue spheres represent elastic matrix of MG. The region in the center with red spheres contains the most loosely packed atoms, and these atoms have higher mobility and shortest intrinsic relaxation time τ_1 , and the orange and yellow sphere regions surrounding the red zone are atoms with lower mobility and

longer intrinsic relaxation time τ_2 and τ_3 ($\tau_2 < \tau_3$). Fig. 1(b) shows the energy distribution of the atoms in the flow unit. The energy from the center of the flow unit to the surrounding harder regions is a Gaussian-like distribution and the energy decreases quickly with the increase of the distance from the center. When the stress or thermal energy is applied, and if the activation time (t), $\tau_1 < t < \tau_2$, the central region with red atoms in the flow unit will be activated; If $\tau_2 < t < \tau_3$, the relative harder orange region will also be activated; and if $t > \tau_3$, the whole flow unit will be fully activated.

Due to the heterogeneous structure of MGs, each flow unit in MG has different average intrinsic relaxation time and activation energy, and the average relaxation time and activation energy of flow units of an MG have a broad distribution [29–31]. The average volume of flow units is $\bar{\Omega} = \Sigma\Omega_i/n$, and the average flow unit activation energy is $W = \Sigma W_i/n$, the average intrinsic relaxation time is $\tau_0 = \Sigma\tau_i/n$, where n is the number of flow units in an MG. So the $\bar{\Omega}$ and W are also a function of the activation time t . Based on the cooperative shearing model [28], the relation of the W and $\bar{\Omega}$ is [28,29]: $W = (8/\pi^2)\xi G\gamma_c^2\bar{\Omega}$, where ξ is a constant, γ_c is the yield strain limit, G is the average shear modulus of the MG, and the relation of $\bar{\Omega}$ and time of the activated flow unit is:

$$\bar{\Omega} = \frac{\pi^2 kT}{8G\xi\gamma_c^2} \ln \frac{t}{\tau_0}, \quad (3)$$

the volume fraction of flow units is:

$$c = n\bar{\Omega}/V = \frac{\pi^2 kTn}{8VG\xi\gamma_c^2} \ln \frac{t}{\tau_0}. \quad (4)$$

Eq. (4) indicates that the density or the volume fraction of the flow units in an MG is activation time dependent.

3. Results

Fig. 2(a)–(c) schematically illustrate time dependence of size and density of flow units in an MG. For short activation time (or high strain rate) as shown in Fig. 2(a), only few and small flow units with higher energy and atomic mobility are activated; with extended time, more and larger flow units are activated as indicated in Fig. 2(b); and if the activation time is long enough, more and more flow units with higher activation energy would be activated [Fig. 2(c)], and when the density of the flow units reaches a critical value the MG will behave like liquids [6,11].

The flow units are found to correlate with properties P such as elastic moduli, hardness, and plasticity of MGs in the form of [21,30–34]: $P = P_m/(1 + \alpha)$, where α correlates to the effective fraction of flow units, and P_m is the property of glass without flow units corresponding to that of perfect crystal or ideal glass [21,31–34]. According to Eqs. (3) and (4), the time dependent activation and fraction of flow units results in the time dependent properties of MGs. We can then investigate the role of time in activation of flow units through the studying of time dependent mechanical properties and macroscopic flow behaviors in MGs.

We used a Sr-based MG to perform the uniaxial compression at room temperature (RT) to observe the yield strength changing under different strain rate ν . The rod of $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Zn}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}$ MG is 2 mm in diameter and 4 mm in length, and has a low T_g of 323 K [35]. The strain rate is equivalent to the activation time t . With a giving ν , the activation time at a given strain γ can be expressed as: $t = \gamma/\nu$. Fig. 2(d) shows that at RT and a higher $\nu = 3 \times 10^{-4} \text{ s}^{-1}$, the MG exhibits an elastic deformation followed by pure brittle behavior, and fractures into small pieces. This indicates that, due to few and small flow units in the MG are activated at RT at high strain rate or shorter activation time, no

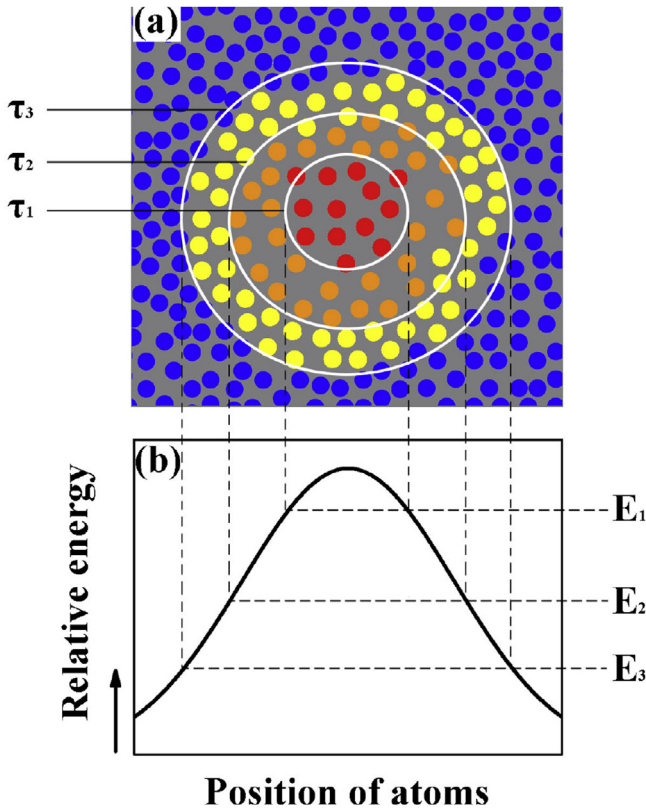


Fig. 1. (a) The schematic illustration of the atomic structure of flow unit in metallic glass. The blue spheres represent the atoms in elastic matrix region, the red spheres represent the atoms in the flow unit with shortest intrinsic relaxation time τ_1 and the orange regions and yellow regions respectively are harder atoms with lower mobility and larger intrinsic relaxation time τ_2 and τ_3 . (b) The energy distribution of the atoms in the flow unit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

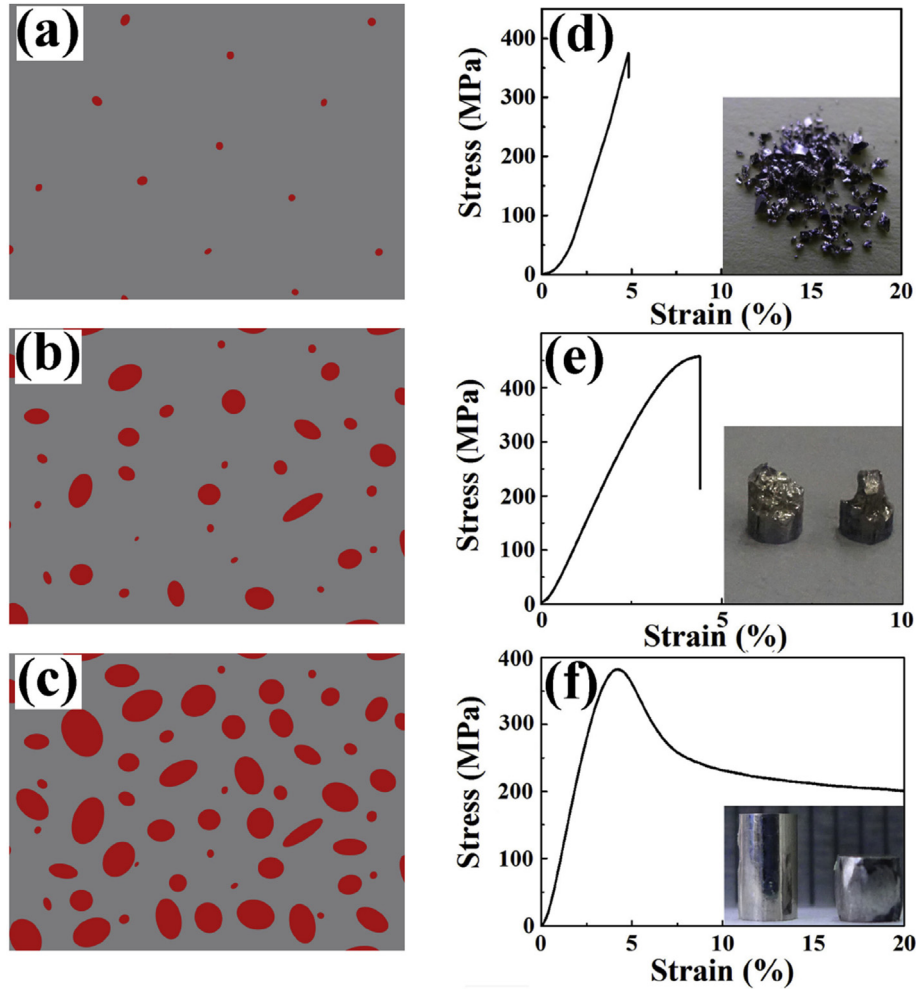


Fig. 2. (a)–(c) The sketches of the flow units activated at different activation times. (d)–(f) the corresponding experimental strain–stress curves of $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Zn}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}$ MG at 3×10^{-4} , 2.5×10^{-4} and 10^{-4} s^{-1} , respectively. (a) and (d) with short activation time; (b) and (e) with normal activation time; (c) and (f) with long activation time. The inserts in (d), (e) and (f) are the photos of the corresponding deformed samples.

plastic deformation occurs. However, when the strain rate decreases or the activation time increases, the MG becomes plastic due to more and more flow units are activated in the MG under the conditions [Fig. 2(e)]. When the ν is below a critical value ($2 \times 10^{-4} \text{ s}^{-1}$) or the activation time reaches a critical value, remarkably, the MG displays homogeneous deformation as larger as 70% [see Fig. 2(f)], which is a liquid like behavior at RT and similar to that observed in supercooled liquid in conventional MGs at high T . The MG transforms into supercooled liquid state at RT realized solely by activating sufficient large fraction of flow units through the sufficient long activation time. The results demonstrate the relationship between activation time of the flow units and macroscopic flow behavior in MGs.

We note that the smaller strength at high strain rate of 3×10^{-4} [see Fig. 2(d)] is due to the very brittle fracture and unstable behavior of the strontium based metallic glass with many casting defects in it. When the strain rate is small, it deforms homogeneously and the casting defects have little effect on it. But when the strain rate is big, the deformation become highly heterogeneous and the defects will lead to the fracture before its intrinsic strength, which is much larger at this strain rate.

The relationship between t and strain γ in a $\text{La}_{75}\text{Ni}_{7.5}\text{Al}_{16}\text{Co}_{1.5}$ MG was also investigated using the mandrel winding method [36]. It is found that with increasing t , the γ increases rapidly at the

beginning, but the rate of increasing γ slows and approaches a saturation value of γ_0 . The changing tendency of the γ with activation time t follows a formula of $\gamma(t) = \gamma_0/(1 + \alpha)$, where α correlates to the fraction of the activated flow units [36]. The results show that for the shorter deformation time, the MG correspondingly shows small γ due to few flow units are activated; With an extension of activation time, more and more flow units are activated, and the MG shows larger γ , which also clearly confirm that the plasticity of an MG is closely correlated with the activation time of flow units [36].

The relation between the activation time of flow units and elastic modulus E in a variety of MGs is studied. The E of the MGs can be roughly expressed as the linear superposition of the Young's modulus of the hard matrix and the flow units as: $E = E_h(1-c) + E_f c$, where E_h and E_f are the modulus of the harder matrix and flow units, respectively, c is the fraction of the activated flow units. Due to E_f of the liquid-like flow units is close to zero, the E of an MG can be expressed as: $E = E_h(1-c)$.

And a linear relationship between the E of an MG and the strain rate ν can be obtained:

$$E = E_h \left(1 - \frac{\pi^2 k T n}{8 G \xi^2 \gamma_c^2} \ln \frac{\gamma}{\tau_0} \right) + \frac{\pi^2 k T n E_h}{8 G \xi^2 \gamma_c^2} \ln \nu. \quad (5)$$

The uniaxial compression is applied to $\text{Ce}_{69}\text{Al}_{10}\text{Cu}_{20}\text{Co}_1$, $\text{La}_{55}\text{Al}_{25}\text{Ni}_5\text{Cu}_{10}\text{Co}_5$ and $\text{Pd}_{40}\text{Cu}_{30}\text{P}_{20}\text{Ni}_{10}$ MGs (2 mm in diameter and 2 mm in length) at $T = (T_g + T_x)/2$ in their supercooled liquid state to test their E changing upon v . Fig. 3 shows the change of E (determined from their strain–stress curves) of the MGs with v (equivalent to the activation time). With the decrease of the v , or equivalent increase of activation time, the E of the three MGs decreases. The strain rate dependent E of MGs, which make MGs behave hardening or softening, is attributed to the increase of fraction of the flow units at a lower strain rate or a longer activation time in MGs, because the MG with more liquid like regions shows larger softening behaviors [21]. The isothermal annealing far below T_g , which can induce annihilation of the flow units, also leads to significant increase of E or stiffer of MGs [32]. As shown in Fig. 3, Eq. (5) can well fit the experiment data of the MGs and confirms that the time dependent flow unit model can describe the change tendency of elastic modulus with the varying of the strain rate. We note that in Fig. 3 the Ce-based bulk metallic glass is much more sensitive to strain rate. This is due to more flow units activated during deformation in Ce-based MG. From Eq. (5), we can see that the slope of the line in Fig. 3 is mainly decided by, the average number of flow units n in metallic glasses.

We further verify the relationship between the activation time and the strength in MGs. The yield strength σ_c of MG is roughly described by an average elastic limit criterion, $\sigma_c = \gamma_c E$, where $\gamma_c = 0.0267$ [28]. From Eq. (5), the relation between σ_c and t (or strain rate v) can be obtained:

$$\sigma_c = \gamma_c E = \gamma_c E_h \left(1 - \frac{\pi^2 k T n}{8 G \xi \gamma_c^2} \ln \frac{t}{\tau_0} \right)$$

or

$$\sigma_c = \gamma_c E_h \left(1 - \frac{\pi^2 k T n}{8 G \xi \gamma_c^2} \ln \frac{\varepsilon}{\tau_0} \right) + \frac{\pi^2 k T n E_h}{8 G \xi \gamma_c} \ln v \quad (6)$$

According to Eq. (6), under a high strain rate, only those flow units with shorter τ_0 can be activated. Contrarily, if the applied strain rate is low or activation time is long enough, the volume fraction of the flow units would increase and finally reach the critical value for yielding. So the yield of an MG can occur in a much lower stress than yield stress with enough long time. In other

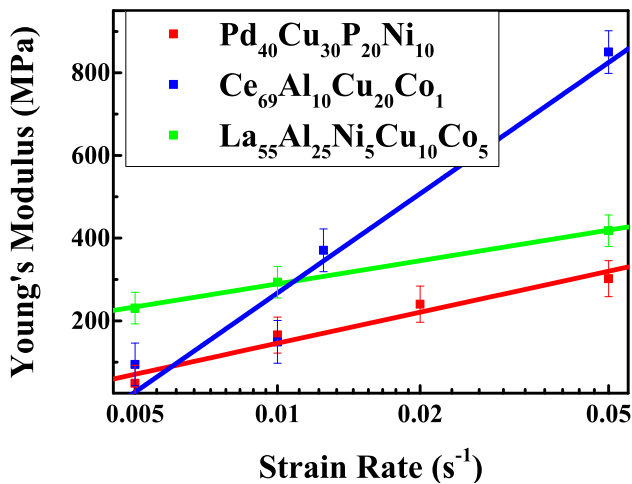


Fig. 3. The elastic modulus of $\text{Pd}_{40}\text{Cu}_{30}\text{P}_{20}\text{Ni}_{10}$, $\text{Ce}_{69}\text{Al}_{10}\text{Cu}_{20}\text{Co}_1$ and $\text{La}_{55}\text{Al}_{25}\text{Ni}_5\text{Cu}_{10}\text{Co}_5$ MGs deformed with different strain rates v . The three lines are the linear fits of the experiment data.

words, the strength of an MG depends on the time dependent fraction of flow units. Fig. 4(a) shows the strain–stress curves of the $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Zn}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}$ MG at RT under different v or activation time t . Due to the low T_g (meaning of lower activation energy of flow units [8]) of the MG, when the v is lower than a critical value, the MG can show pronounced homogeneous deformation in limited applied time and its maximum strength σ_c decreases with the decrease of the v (or increase of the activation time) as clearly shown in Fig. 4(b), which is in agreement with the results of Refs [24–27]. It is shown that there is a linear relationship between σ_c and $\ln t$ in Fig. 4(b). The fit of Eq. (6) in Fig. 4(b) has a perfect accordance with the experiment data confirming that time-dependent flow units strongly affect the strength of MGs.

4. Discussions

We note that the activation time t is equivalent to the observation time t_0 for the flow units [9,36–39]. According to the elastic model [9,39], for a flow unit with intrinsic relaxation time τ_0 , if the t_0 is very long, one can see the flow unit flows. On the other hand, if τ_0 of a flow unit is much longer than t_0 , the flow unit is solid-like. This means that any flow unit is solid-like when it is probed on a sufficiently short time scale, and its short time elastic properties are characterized by the instantaneous elastic moduli of G_∞ and K_∞ [9]. In other words, the viscosity or dynamic in flow unit is determined by the observation time or activation time, and this is the intrinsic reason for the time dependent properties of the MG. When the observation time scale is changed, the density, size and activation energy of flow units are different as illustrated in Fig. 2. If the observation time scale is short enough, the fraction of the activated flow units are so small that the MG just shows elastic behavior. If the observation time scale is long enough, the fraction of the activated flow units become so big that the MG shows liquid like behavior as shown in Fig. 2(f).

A diagram of the flow (for both plastic deformation and glass to liquid transformation) of an MG therefore can be constructed based on the concept of flow units, activation time t , and imposed energy E (stress or temperature) as illustrated in Fig. 5. The flow of an MG depending on applied energy E and activation or observation time t due to the density and size of flow units connect with activation time. If the activation time is short, few flow units are activated, and the MG behaves solid like. If t is longer than the τ_0 of flow units, which has a broad distribution in temporal scale, or high enough energy E is applied, a greater number of flow units will be activated. When the density of flow units achieves a critical point, or $E > E_{FU}$ (the activation energy of the flow units), the percolation of flow

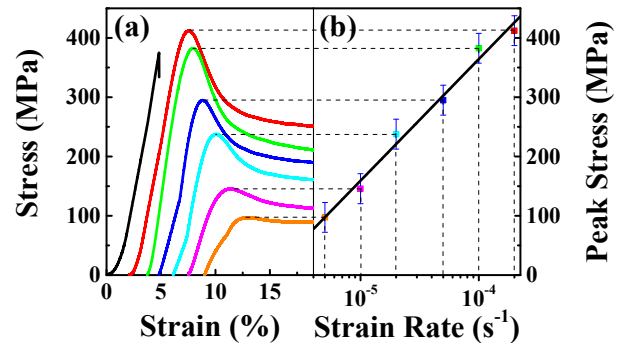


Fig. 4. (a) The strain–stress curves of $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Zn}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}$ MG with different strain rates: from left to right are 3×10^{-4} , 2×10^{-4} , 1×10^{-4} , 5×10^{-5} , 2×10^{-5} , 1×10^{-5} and 5×10^{-6} s^{-1} respectively. (b) The peak stresses of the strain–stress curves at different strain rates. The black line is the linear fit.

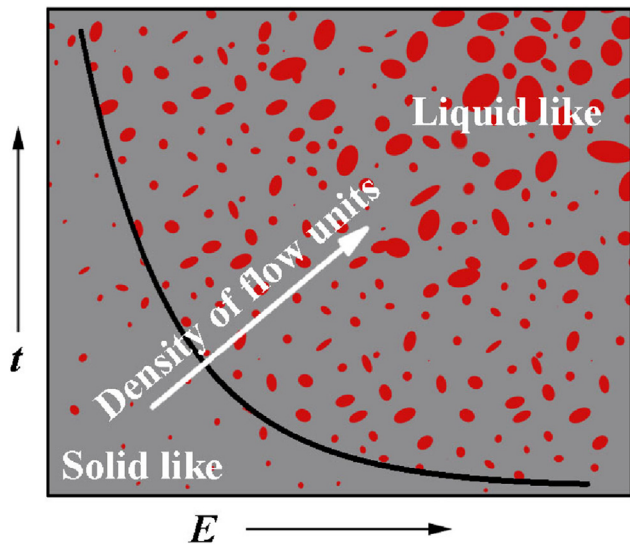


Fig. 5. A schematic illustration of the relationship between flow units, activation time t and the applied energy E (stress or temperature). The background is a sketch of the activated flow units in MGs. The arrow shows the flow units increase with the observing time or the applied energy. The black line is a dividing line between the liquid like and the solid like behaviors in MGs.

units occurs and leads to a homogeneous plastic deformation or glass to liquid transition depending on the applied energy which could be stress or temperature. The diagram also implies that if the $t > \tau_0$ or if enough energy is applied to activate a sufficiently high density of flow units, the macroscopic flow can occur in any MG. This is similar to that of a jammed system, which has transitions with the changing of temperature, stress or density [40]. Our previous work [37] shows that the observation time, equivalent to temperature and stress, is key parameter to a glass transition diagram in metallic glasses, and a three dimensional diagram involved in the observation time, stress and temperature is obtained to describe the glass transition in MGs [37]. The temperature, observation time, and stress are equivalent control parameters, and the change of any of them has equivalent role in glass transition or plastic deformation. Due to the stress and temperature are considered as equivalent in the energy point of view [5], the three dimensional diagram can degenerate into the two dimensional diagram of Fig. 5. The time dependent flow units model provide insight on the structure origin for the 3D diagram of glass to supercooled liquid transition involved in the observation time, stress and temperature in glasses. The result of the time dependent of flow units herewith can also help clarify the ambiguities in estimating the shear transformation sizes or flow units by different studies [41–45], because the different technique methods or simulations involve in different time scale or different applied energy.

5. Conclusions

We show how the flow units in metallic glasses are activated with time, which leads to the time dependence of the features and properties of MGs. We use uniaxial compressions with different strain rates to illustrate the relationship between the activation time and the flow units. It is found that the size and the fraction of

the flow units increase with the applied time, which lead to the time dependences of the mechanical behaviors, flow phenomenon and relaxations of MGs. With different activation time, the MG shows markedly different mechanical behaviors; if the strain rate is large enough, a supercooled liquid or even a common liquid may behave like a solid with an amazing strength, and if the strain rate is small enough, a glass can behave like a liquid showing homogeneous flow. These experimental observation are in accordance with our time dependent flow unit model. A diagram for the flow in glass based on the concept of flow unit, activation time, and imposed energy (stress or temperature) is constructed to understand the deformations and flow mechanisms of MGs.

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